A highly sensitive pressure sensor using conductive composite elastomers with wavy structures

Rujie Sun[a], Xiao-Chong Zhang[a], Jonathan Rossiter[b], Fabrizio Scarpa[a*]
[a]Advanced Composites Centre for Innovation and Science, University of Bristol, Queen's Building, Bristol, BS8 1TR, UK; [b]Department of Engineering Mathematics, University of Bristol, Merchant Venturers Building, Bristol, BS8 1UB, UK.

ABSTRACT

Flexible pressure sensors are crucial components for the next generation wearable devices to monitor human physiological conditions. In this paper, we present a novel resistive pressure sensor based on hybrid composites made from carbon nanotube (CNT) for the conductive coating layer and polydimethylsiloxane (PDMS) elastomers as the substrate. The high sensitivity of these sensors is attributed to the change of contact resistance caused by the variation of the contact areas between the wavy film and the electrodes. Porous electrodes were designed to increase the roughness of the interfaces, thus further enhancing the pressure sensitivity. The developed device was verified through a series of tests, and the sensor exhibited a high sensitivity of 2.05 kPa^{-1} under a low pressure of 35.6 Pa,

Keywords: pressure sensor, contact resistance, wavy structure, porous electrodes, nanocomposites

1. INTRODUCTION

Recently, significant advances have been achieved in wearable electronics by introducing multifunctional properties, such as flexibility, stretchability, injectability, and biocompatibility [1]. As an essential part of wearable electronics for human physiological monitoring, pressure sensors are drawing increasing attention. The current research activities can be categorised into three main areas: advanced materials, intelligent structures, and novel manufacturing approaches. Nanomaterials with excellent electrical properties show great potential for sensing applications, such as carbon nanotubes (CNTs) and graphene. CNTs exhibit distinct features, including excellent conductivity and high strain gauge. Bao et al [2] developed skin-like pressure and strain sensors based on transparent elastic films of CNTs. Spring-like structures of CNTs were fabricated by axial pre-strain and subsequent release accommodating up to 150% stretchability with high conductivity. Graphene also shows great electrical and mechanical properties due to its unique two-dimensional hexagonal structure, which makes it relatively flexible in the through-thickness direction. Various concepts have been evaluated to achieve highly sensitive strain sensors. Human hair is a mechanically strong and flexible fibre that is a promising candidate as a strain sensor substrate. With reduced graphene oxide coated on its surface, hair-based sensors can detect various deformations including stretching, bending, and compression [3]. Novel graphene configurations can represent another efficient way to improve the sensitivity and reversibility, such as graphene woven fabrics with crisscross interlaced ribbons [4, 5] and graphene-based fibres with compression spring architecture [6].

A sensor is a device that detects and converts a mechanical form of energy to relevant electrical signals based on various transduction mechanisms, including traditional methods (i.e. piezoresistivity [7, 8], piezoelectricity [9, 10], capacitance [11, 12]) and recently developed ones (triboelectricity [13-15], optics [16]). The piezoresistive effect is a resistance change induced by an applied mechanical strain. Piezoresistive interlocked microdome arrays are employed in a tactile-direction-sensitive sensor, which could distinguish various mechanical stimuli due to the different deformation levels of microdome architecture under different directional forces [7]. The pyramidal microstructure is another efficient topology to achieve a highly tuneable resistance pressure sensor [8]. An elastic pyramidal microstructured polydimethylsiloxane (PDMS) is spray-coated with a layer of single-wall carbon nanotubes (SWNTs), and the height of the SWNT coating is controlled to tune the threshold of resistance switching range. The piezoelectric effect is a reversible process between electrical charge and external stimuli. It exhibits high sensitivity and rapid response time when converting mechanical signals to electrical signals. A bioinspired interlocked geometry with hierarchical micro-

[*] Corresponding author: E-mail f.scarpa@bristol.ac.uk; Phone: +44 (0) 117 33 15306.

and nano- structured ZnO nanowire arrays has been developed for the detection of both static and dynamic tactile stimuli based on piezoresistive and piezoelectric transductions [9]. The capacitive sensor, normally comprising two electrodes and dielectric layer, is a capacitor with variable capacitance. Considerable attention has been drawing on the designs of dielectric structure. Bao and co-workers demonstrated various microstructured dielectrics that could significantly improve the sensitivity of pressure sensors, including a pyramidal-shaped dielectric layer integrated with a signal amplification component based on microhair structures [12], hybrid structure with porous PDMS and air gap as dielectric layer to keep high sensitivity in both low and high pressure regions [17]. Recently, other novel transduction methods have attracted significant interest for sensing performance improvement. Triboelectricity based on the triboelectric effect coupled with electrostatic induction shows potential in self-powered sensing systems [18]. The micropyramid-featured pattern is also demonstrated to be able to induce large triboelectric charge density, thus enhancing the performance [13, 15]. In addition, an alternative approach based on polymer waveguides without any electronic components in sensing areas is proposed for dynamic response detection [16]. The waveguides underneath the touch layer would scatter upon a pressure touch, and by monitoring the light intensity the force could be detected.

In this paper, we describe the development of a resistive sensor to enhance the pressure sensitivity by combining the properties of CNTs with a structure having a topological waviness. The high sensitivity is attributed to the variation of contact resistance at the interfaces of the wavy film and the electrodes. PDMS polymer was used as the substrate of the wavy structure due to its biocompatibility and its excellent flexibility. CNTs were then coated onto the surface of the wavy film to create a sensitive and conductive layer. Upon an external pressure, the geometrical change of the wavy structure is induced, resulting in an increase of the contact areas between the wavy film and the electrodes, thereby leading to a decrease in resistance and improving the pressure sensitivity of the sensor. With the release of the pressure, the wavy structure would recover to its initial state due to the excellent elasticity of PDMS. More noteworthy is the design of the electrodes that used porous foam as the substrate and CNT coating was deposited on the foam surface. The flexibility and rough surface of the foam-based electrode made the contact resistance more sensitive to the applied pressure.

2. EXPERIMENTAL

2.1 Material synthesis

Conductive ink: Carboxyl group (-COOH) functionalized Multi Walled Carbon Nanotubes (MWNTs, CheapTubes) with diameters of 10~20 nm and lengths 10~30 μm were used as the base of the conductive ink and dispersed into Isopropyl alcohol (IPA) solution. Polyvinylpyrrolidone (PVP) from Sigma-Aldrich was adopted before sonication as the dispersant. 70.74 mg MWNTs was firstly dispersed in 30 ml IPA, and then 30 mg PVP was added to suspend carbon nanotubes. The ink was then stirred for 5 min, and finally sonicated for 10 min using an ultrasonication probe at 40% amplitude. The final ink contains 0.3% of MWNTs by weight.

Silane solution: Silane was used as the coupling agent of CNTs and the PDMS film. The prepared solution has 0.5 wt % of silane (Sigma-Aldrich). 50 ml of IPA was prepared, and its PH value was adjusted between 3.5 and 4 by adding acetic acid. Then, 58.6 μl of silane was added to the above solution, and stirred for 5 min.

2.2 Resistive pressure sensor fabrication

Conductive wavy film: PDMS base and its cross-linker (Sylgard 184, Tow Corning) was mixed (10:1 w/w) and stirred for 10 min. After degassing under vacuum for 30 min, the mixture was cast into a Perspex wavy mold and then cured at 85 ºC for 1 h before it was peeled off from the mold. The wavy PDMS film was washed by DI water, and dried in the oven at 85 ºC for 10 min. Then, it was treated with O_2 plasma (2 min). After the above steps to render it hydrophilic, the film was immersed in silane solution for 10 min and dried again and a silane layer was created on its surface. The film was then immersed into the conductive ink for 5 min and dried at 85 ºC for 10 min, and this process was repeated 5 times. Finally,a uniform MWNT layer was deposited onto the surface of the wavy film.

Fabrication of electrodes: Porous polyurethane foam (SM Upholstery, open cell, 27kg/m^3 with 2047-2244 pore/m) was used as the substrate of the electrodes. The foam was firstly treated with O_2 plasma (1 min). Then, a similar process as described above was conducted to create a silane layer on its surface. After that, the foam was immersed into the conductive ink for 2 min and dried at 85 ºC for 10 min. In this way, an electrode with rough surface was created.

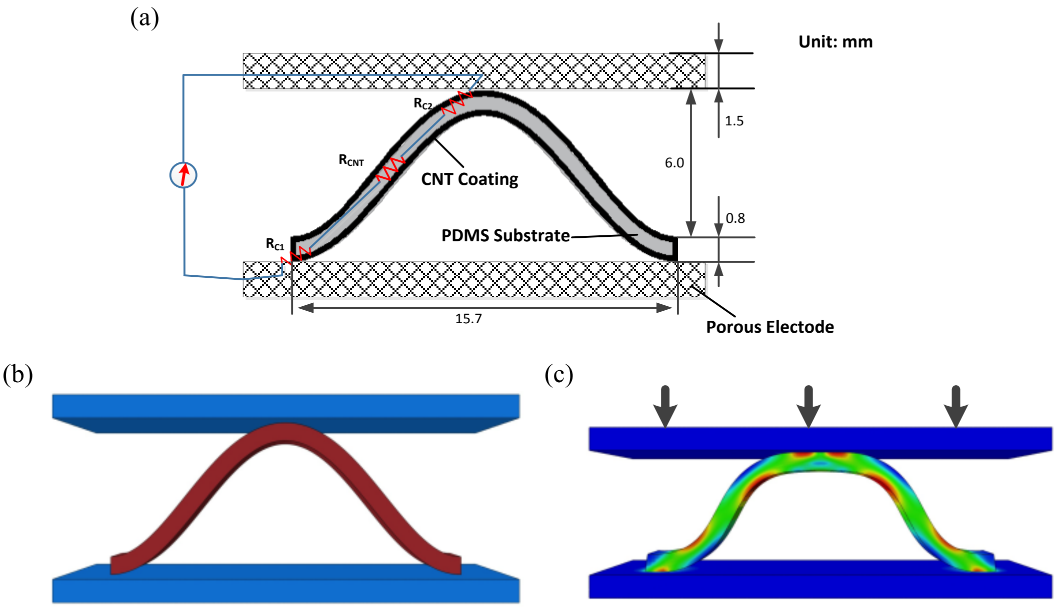

Figure 1. Schematic illustration of the sensor structure. a) diagram of the working mechanism of the sensor with a simplified circuit; b) and c) the variation of the contact areas at the interfaces between the wavy film and the electrodes before and after an applied pressure.

2.3 Characterization

The surface morphologies of the wavy films and foam-based electrodes were characterised by scanning electron microscopy (JSM-6330F). For the real-time pressure-resistance measurement, a measuring system was set up with the resistance measured by Keithley 2100 source meter, and a custom-coded LabVIEW program being used to record the time domain signals. A Canon camera was used to capture the shape changes of the wavy film as well as the variation of the contact areas between the wavy film and the electrodes.

3. RESULTS AND DISCUSSION

3.1 Device structure and working mechanism

Fig.1 illustrates the design concept and working principle of the pressure sensor. It consisted of two key components: the electrodes and the resistive element. Porous foam was used as the substrate of the electrode, and CNTs were coated on its surface to make it conductive. The wavy CNT-coated PDMS film was considered as the resistive element. To better understand the working mechanism of the pressure sensor, a simplified circuit was developed, as shown in Fig. 1(a). In this circuit, the resultant resistance (R) was composed of three parts: namely contact resistances R_{C1} and R_{C2} at the interfaces of the wavy film and top/bottom electrodes, and the resistance of the wavy film R_{CNT}. Applying an external pressure would easily increase the contact areas between the wavy film and the electrodes, thus reducing the contact resistance. Meanwhile, the external pressure also reduced the effective length of the resistive element, thus resulting in a reduction of the ohmic resistance of the sensor. This wavy shape would induce a large variation of contact resistance even under a low pressure. In addition, the rough surfaces of the CNT-coated PDMS film and porous foam further enhanced the sensitivity of the sensor. The electrodes and wavy films were then characterized in detail by using the scanning electron microscopy (SEM) images. Fig. 2 shows the surface morphologies of PDMS film before and after CNT ink coating at two different scales. As shown in figure Fig. 2(a) and (b), the smooth surface can be observed before coating. However, after the CNT coating the presence of some random cracks was clearly observed at a large scale (Fig. 2(c)). Further observations indicate that at micoscale view the CNTs were uniformly deposited on the surface with individual CNT observed (Fig. 2(d)). Fig. 3 shows the surface morphology of the porous foam. The porosity can be clearly seen in Fig. 3(a), and CNTs were also uniformly coated on the foam electrodes, as shown in Fig. 3(b). The advantage of a uniform CNT coating was the establishment of a continuous conductive network, thus avoiding any sudden changes in resistance upon an external pressure. The cracks observed in Fig. 2(c) and 3(b) were caused by the silane layer during the drying process.

(a)

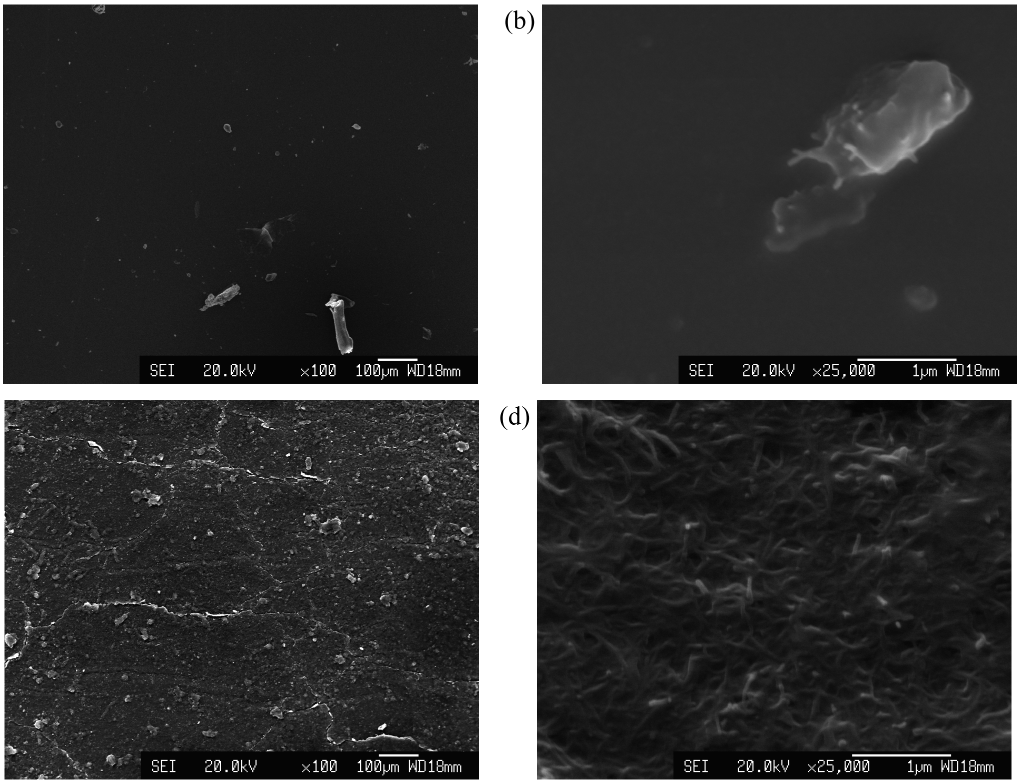

Figure 2. The surface morphologies of the PDMD film. a) and b) SEM images of PDMS film before CNT coating under magnifications of 100X and 25,000X respectively; c) and d) SEM images of PDMS film after CNT coating under magnifications of 100X and 25,000X respectively.

(a)

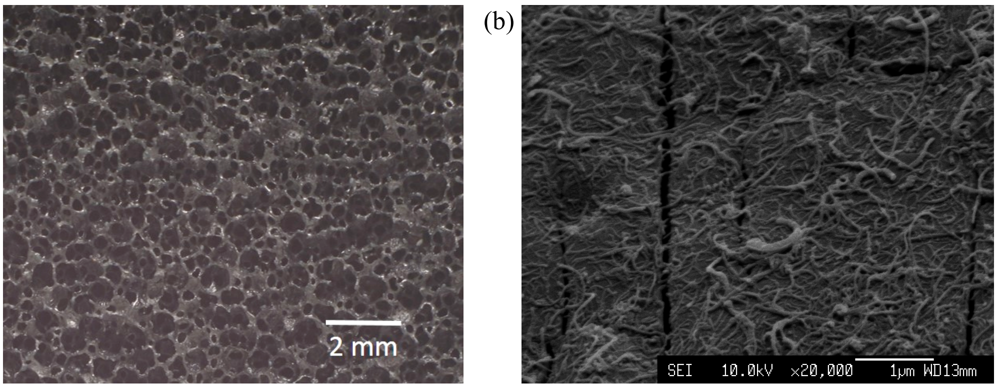

Figure 3. The surface morphologies of the porous foam. a) images (Canon Eos-7D camera) shown the porosity of the foam surface; b) SEM images of the uniform CNT coating.

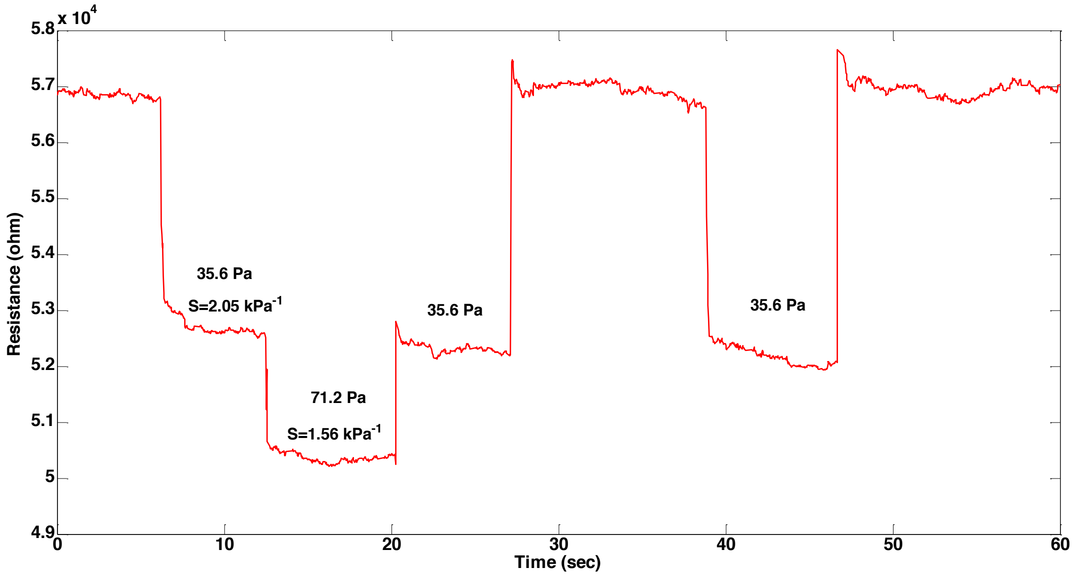

Figure 4. The change in resistance of the pressure sensor over time under real-time testing with different weights on the devices.

3.2 Measurement of pressure sensitivity

To demonstrate the performance of the developed device, pressure testing was conducted. A weight stacking (Perspex, 655mg) with a cross section area 18×10 mm^2 was employed to uniformly apply the pressure on the sensor, generating pressure of 35.6 Pa per single stack. Specifically, the pressure sensitivity is defined as follows:

$$S = \frac{\delta\left((R_p - R_0)/R_0\right)}{\delta P} \quad (1)$$

Where R_p and R_0 are the resistance with and without the pressure respectively, and P is the applied pressure. Fig. 4 shows the change in resistance observed under the applications of two pressures, 35.6 Pa and 71.2 Pa. The sensitivity exhibited a high value of 2.05 kPa^{-1} under a low pressure of 35.6 Pa, whereas the sensitivity decreased to 1.56 kPa^{-1} when the pressure was increased to 71.2 Pa. This phenomenon can be explained by the fact that the contact is more sensitive at first due to the roughness of the interface, and the anisotropic deformation of the wavy shape. Moreover, upon the release of the applied pressure, the resistance would return to its initial value with a fast response time (~30 ms).

4. CONCLUSION

In this work, we describe the development of a new type of pressure sensor based on a conductive wavy structure. The wavy film was prepared by coating a layer of CNTs on a wavy PDMS substrate, serving as the sensitive component of the resistive sensor. The high sensitivity was attributed to the contact resistance at the interface between the wavy film and the electrodes, as well as the piozeresistance of the wavy film. By coating a layer of CNTs, a conductive porous foam was fabricated for the electrodes. The porosity and irregularity of the electrodes could further enhance the sensitivity of the sensor, especially for the detection of low levels of pressure. The proposed sensor exhibited a maximum value of pressure sensitivity of 2.05 kPa^{-1} under a pressure of 35.6 Pa, and the signals were repeatable and stable.

ACKNOWLEDGEMENTS

The authors would like to acknowledge the support of the Engineering and Physical Sciences Research Council through the EPSRC Centre for Doctoral Training in Advanced Composites for Innovation and Science (grant number EP/L016028/1). Jonathan Rossiter is EPSRC Fellow (grant number EP/M020460/1) and also supported by the RoboSoft Coordination Action on Soft Robotics (FP7). Rujie Sun is also grateful to acknowledge the support of the China Scholarship Council.

REFERENCES

[1] J. A. Rogers, "ELectronics for the human body," JAMA 313, 561-562 (2015).
[2] D. J. Lipomi, et al., "Skin-like pressure and strain sensors based on transparent elastic films of carbon nanotubes," Nat Nano 6, 788-792 (2011).
[3] W. Yuan, Q. Zhou, Y. Li, G. Shi, "Small and light strain sensors based on graphene coated human hairs," Nanoscale 7, 16361-16365 (2015).
[4] Y. Wang, et al., "Wearable and Highly Sensitive Graphene Strain Sensors for Human Motion Monitoring," Adv. Funct. Mater. 24(29), 4666-4670 (2014).
[5] T. Yang, et al., "Tactile Sensing System Based on Arrays of Graphene Woven Microfabrics: Electromechanical Behavior and Electronic Skin Application," ACS Nano 9(11), 10867-75 (2015).
[6] Y. Cheng, R. Wang, J. Sun, L. Gao, "A Stretchable and Highly Sensitive Graphene-Based Fiber for Sensing Tensile Strain, Bending, and Torsion," Adv. Mater. 27(45), 7365-7371 (2015).
[7] J. Park, et al., "Tactile-Direction-Sensitive and Stretchable Electronic Skins Based on Human-Skin-Inspired Interlocked Microstructures," ACS Nano 8(12), 12020-12029 (2014).
[8] H.-H. Chou, et al., "A chameleon-inspired stretchable electronic skin with interactive colour changing controlled by tactile sensing," Nat. Commun. 6, 8011 (2015).
[9] M. Ha, et al., "Bioinspired Interlocked and Hierarchical Design of ZnO Nanowire Arrays for Static and Dynamic Pressure-Sensitive Electronic Skins," Adv. Funct. Mater. 25(19), 2841-2849 (2015).
[10] C. Dagdeviren, et al., "Conformal piezoelectric systems for clinical and experimental characterization of soft tissue biomechanics," Nat. Mater. 14, 728-736 (2015).
[11] S. C. B. Mannsfeld, et al., "Highly sensitive flexible pressure sensors with microstructured rubber dielectric layers," Nat. Mater. 9, 859-864 (2010).
[12] C. Pang, et al., "Highly Skin-Conformal Microhairy Sensor for Pulse Signal Amplification," Adv. Mater. 27(4), 634-640 (2015).
[13] Y. Yang, et al., "Human Skin Based Triboelectric Nanogenerators for Harvesting Biomechanical Energy and as Self-Powered Active Tactile Sensor System," ACS Nano 7(10), 9213-9222 (2013).
[14] Y. Aifang, et al., "Triboelectric sensor as self-powered signal reader for scanning probe surface topography imaging," Nanotechnology 26(16), 165501 (2015).
[15] F.-R. Fan, et al., "Transparent Triboelectric Nanogenerators and Self-Powered Pressure Sensors Based on Micropatterned Plastic Films," Nano Lett. 12(6), 3109-3114 (2012).
[16] S. Yun, et al., "Polymer-Waveguide-Based Flexible Tactile Sensor Array for Dynamic Response," Adv. Mater. 26(26), 4474-4480 (2014).
[17] S. Park, et al., "Stretchable Energy-Harvesting Tactile Electronic Skin Capable of Differentiating Multiple Mechanical Stimuli Modes," Adv. Mater. 26(43), 7324-7332 (2014).
[18] Z. Wang, J. Chen, L. Lin, "Progress in triboelectric nanogenerators as a new energy technology and self-powered sensors," Energy Environ. Sci. 8(8), 2250-2282 (2015).